

# ALUMINUM OR ALUMINUM ALLOY REFINING METHOD

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## Abstract of JP55050442

PURPOSE: To refine molten Al contg. metallic impurities by adding a specified amt. of Mn and Mg to the Al after which they are melted and cooled to a specified temp. to crystallize the impurities as intermetallic cpds., which are then separation-removed. CONSTITUTION: To molten Al or a molten Al alloy contg. metallic impurities such as Fe, Cr, Mo, Pb and As are added metallic Mn or an Al-Mn alloy contg. 0.6-6 times as much Mn as the impurities and metallic Mg or an Al-Mg alloy contg. 0.002-0.1 time as much Mg as the molten metal. They are melted, and the mixed molten metal is cooled and held at the eutectic point to a temp. 50 deg.C above the point. Thus, the impurities are crystallized as intermetallic cpds. with Al-Mn type metal. The crystallized cpds. are then separated by filtration of other method. By this method high purity Al or a high purity Al alloy is recovered.

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⑯ アルミニウム又はアルミニウム合金の精製法

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## 明細書

## 1. 発明の名称

アルミニウム又はアルミニウム合金の精製法

して鉄その他クロム、モリブデン、鉛、ヒ素等の金属不純物を効率的に除去して Al を精製する方法に関するものである。

## 2. 特許請求の範囲

(1) 金属不純物を含有するアルミニウム又はアルミニウム合金浴湯中に、該浴湯に含まれる金属不純物の 0.5 ~ 6 倍量に相当するマンガンを含む金属マンガン及び/又はアルミニウム—マンガン合金と、該浴湯の 0.002 ~ 0.1 倍量に相当するマグネシウムを含む金属マグネシウム及び/又はアルミニウム—マグネシウム合金を添加して溶融混合し、この混合浴湯を、その共晶温度乃至それより 50°C 高い温度の範囲まで冷却して保持し、金属不純物を金属間化合物として晶析分離することを特徴とするアルミニウム又はアルミニウム合金の精製法。

## 3. 発明の詳細な説明

本発明はアルミニウム又はアルミニウム合金(以下これらをまとめて Al と表示することがある)の精製法に関し、詳細には Al 中に含まれる主と

近年のアルミニウム産業においては、省資源及び地金価格の安定等のために Al スクラップの有効利用が検討されている。ところが Al スクラップには鉄をはじめとする多くの金属不純物が含まれており、その精製技術が確立されておらないから、一部鉄物用合金の原料として使用されている程度で、比較的成分規制の厳しい延伸材用合金の原料としては殆んど利用されていない。

これらから Al の精製法について種々の研究が進められ、④ Al 中の金属不純物を電解により分離する 8 層式電解精製法、⑤ 金属不純物特に鉄を Al—珪素—鉄の金属間化合物として分離する方法、⑥ Al 浴湯中に高融点の Al—マンガン合金を添加して該固相合金に不純物特に鉄を吸着させ、Al—マンガン合金との金属間化合物として除去する方法、等が提案されている。ところが前記④の方法では初めて高純度の Al が得られる反面、精製作業が煩雑で且つ精製費用が極めて高く、ス

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タフツアの精製処理法としては不適当である。また⑥の方法では精製工程でA<sub>l</sub>中に多量の珪素が混入するから、その用途は極めて特殊なA<sub>l</sub>-硅素合金に限定される。これらに比べて前記⑤の方法は、精製作業が簡単で鉄を相当低減することができ、またA<sub>l</sub>中に少量のマンガンが混入することがあつてもその不純物効果が殆んど表われないから、工業性の点では最も好ましい方法といふことができる。しかしこの方法は固・液反応によつて鉄を吸着除去しようとするものであるから、鉄とA<sub>l</sub>-マンガン合金との接触頻度が少なく且つ反応速度も極めて遅い。そのため鉄の除去率は他の方法に比べて相当低く、成分規制の厳しいA<sub>l</sub>を得るための精製法としての利用価値は乏しい。

本発明者等は前述の様な事情のもとで、特に前記⑤の方法に着目し、A<sub>l</sub>-マンガン合金をA<sub>l</sub>と溶融状態で接触させれば金属不純物との接触頻度及び反応速度が高められ、不純物除去効率を向上できると考え、この着想を実現すべく研究に着手した。その結果、前記⑤法即ち固・液接触反応

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に比べれば除去効率を相当改善できるが、依然満足し得るまでには至らなかつた。ところがその後更に研究を重ねた結果、精製すべきA<sub>l</sub>中にマンガンと共に適量のマグネシウムを添加し、得られる溶融混合物をその共晶温度より若干高い温度まで冷却してやれば、金属不純物の殆んどがA<sub>l</sub>-マニシアン系金属との金属間化合物として晶析分離されることを知つた。しかもこの工程で鉄はもとより、クロム、モリブデン、鈴、ヒ素等の不純物も効率良く除去できることを知り、茲に本発明を完成するに至つた。

即ち本発明に係る精製法とは、金属不純物を含有するA<sub>l</sub>又はA<sub>l</sub>合金溶湯中に、該浴湯に含まれる金属不純物の0.5~6倍量に相当するマンガンを含む金属マンガン及び/又はA<sub>l</sub>-マンガン合金と、該浴湯の0.002~0.1倍量に相当するマグネシウムを含む金属マグネシウム及び/又はA<sub>l</sub>-Mg合金を添加して溶融混合し、この混合浴湯を、その共晶点乃至それより50°C高い温度の範囲まで冷却して保持し、金属不純物をA<sub>l</sub>-

(4)

マンガン系金属との金属間化合物として晶析させて分離するところに要旨が存在する。

以下図面によりながら本発明の構成及び作用効果を詳細に説明するが、下記は代表例にすぎず、前・後記の趣旨に従して適宜に変更して実施することも可能であり、それらはすべて本発明技術の範囲に含まれる。

第1図はA<sub>l</sub>-マンガン(Mn)-鉄(Fe)系の8元状態図であり、ABC'Dの領域では初晶として(Fe,Mn)Al<sub>6</sub>の金属間化合物が生成する。ここでX点で示される組成の浴融混合物に対して、Y点で示される組成になる如くA<sub>l</sub>-Mn合金を添加して完全に浴融させた後、この合金の液相線温度以下で且つ共晶温度以上のある温度まで冷却して保持すると、(Fe,Mn)Al<sub>6</sub>の金属間化合物が晶出し、浴融物はZ点で示される組成に変化する。従つてこの晶析物を分離することによつてFe濃度の低い浴融A<sub>l</sub>合金を得ることができる。

しかしながらこの方法ではFeの除去が尚不十分

(5)

分で且つA<sub>l</sub>合金中へのMnの浴存量も相当大きい。ところが、A<sub>l</sub>-Mn合金の添加と同時にC或はA<sub>l</sub>-Mn合金が完全に浴融した後浴湯を冷却する過程でA<sub>l</sub>-Mg合金を添加すると、8元状態図はたとえば第1図の頂極で示す状態に変化し、(Fe,Mn)Al<sub>6</sub>の晶出領域はAB'C'Dとなつてより低温側に広がる。即ち浴融混合物中に更にA<sub>l</sub>-Mg合金を共存させることによつて共晶温度が低下し、より低温で(Fe,Mn)Al<sub>6</sub>を晶析させることができると、Z点をZ'点まで移動させることができなり、この晶析物を同様に分離除去することによつてA<sub>l</sub>中のFe及びMnの除去率を大幅に高めることができる。

尚第2図はMgの添加効果を示す他の例で、晶析物の分離除去前・後におけるA<sub>l</sub>合金中のFe及びMnの含有率を示すグラフである。この結果からも明らかに如く、Mg無添加ではFe及びMnの含有率をH線(第1図のB-C線に対応)相当量まで低減するのが限度であるが、Mgを8%添加するとI線相当量まで、Mgを6%添加すると

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J線相当量まで、Mgを10%添加するとE線相当量まで、Mgを20%添加するとL線相当量まで、Fe及びMnの含有率を低下することができる。この様に本発明では不純物として鉄を含むAl溶湯に對してMnと共にMgを添加することにより、またMgの添加量を増加することによつて、Fe及びMnの含有率を大幅に低下させることができる。また後記実施例でも明確にするが、本発明によればFeのみならず、クロム、モリブデン、鉛、ヒ素等の金属不純物についても大幅に低減できることがわかつた。

尚第8図は86%Al-2%Mn-2%Feに對するMgの添加率と共晶温度(融点)の関係を測定した結果を示すもので、Mgは被処理Al材の融点を低下させるうえでも卓効を示す。

本発明で使用されるMnは、金属Mn及びAl-Mn合金の何れの形で用いてもよいが、Al中に添加したとき可及的速やかに溶解させるためには、5~80%程度のMnを含むAl-Mn合金が最も望ましい。またMnを添加するときのAl

(7)

溶湯の温度は、溶融速度を加味して720~850°Cが最も好ましい。このときのMnの添加量は、処理すべきAl溶湯中に含まれるFe量に對してMn純分として0.5~6.0倍量の範囲から選択すべきである。しかしてMn量が前記範囲では金属間化合物の量が不足してFeその他の金属不純物を十分に除去することができず、一方前記範囲を越えると、第4図から明らかのように不純物除去率がほとんど上向せず、また晶出する(Fe, Mn)Al<sub>6</sub>の量が増大する結果Al精製物の量が減少し、歩留りが低下するので実用的でない。

またMgも金属Mg或はAl-Mg合金の何れの形で用いてもよくその添加温度は、Al-Mn合金を添加するときの温度よりもやや低いめ(Al-Mg合金の方がAl溶湯中に溶解し易い温度)の700~800°Cの範囲が最適である。ここでMgの添加量は、処理すべきAl溶湯全量に對して0.002~0.1倍量の範囲から選択しなければならない。しかして前記範囲では共晶温度低下効果が殆んど表われず、Feをはじめとする金

(8)

格不純物及びMnの含有率を十分低下させることができず、一方前記範囲を越えると、第5図から明らかのようにFe, Mnはじめ金属不純物の除去率がそれほど顕著でなくなり、本発明の目的を達成し得なくなるからである。

この様にしてAl溶湯中に適量のMn(又はAl-Mn合金)とMg(又はAl-Mg合金)を加えて溶解融合し、これを徐々に冷却することによつて(Fe, Mn)Al<sub>6</sub>を晶析させるのであるが、溶湯の温度が液相線温度以上では(Fe, Mn)Al<sub>6</sub>が晶出せず、また共晶温度以下になると(Fe, Mn)Al<sub>6</sub>と共にAl純品も晶出するから、(Fe, Mn)Al<sub>6</sub>のみの分離は不可能である。従つて晶析温度は当然共晶温度以上液相線温度以下の範囲にすべきであるが、本発明者等が、金属間化合物の晶析による金属不純物の除去率を最大限に高めるべく晶析温度を逐々検討したところ、その共晶温度以上乃至それより50°C高い温度の範囲が最適であり、この温度範囲内で保持することによつて(Fe, Mn)Al<sub>6</sub>の晶析を最も効率よく行なえることが

(9)

確認された。

尚晶析した金属間化合物を溶融Alから分離する手段は特に限定されず、公知の方法或は今後開発されるであろうすべての固・液分離法を適用することができ、具体例としては、金属間化合物の比重が溶融Alの比重よりも大きいことを利用した沈降分離法、多孔質材による沪過分離法、遠心分離法等が代表的なものとして挙げられる。

本発明は概略以上の様に構成されており且つ実施されるが、要は金属不純物を含むAl溶湯に對して適量のMn及び/又はAl-MnとMg及び/又はAl-Mgを添加し、所定温度で金属間化合物を晶析させる方法を採用することにより、Al溶湯中のFeをはじめとするクロム、モリブデン、鉛、ヒ素等を擰めて効率的に除去して高純度のAl又はAl合金を回収し得ることになつたもので、Alスクラップをはじめとする種々の不純なAlやAl合金の精製法としての実用価値は絶る大きい。

次に本発明の実施例を示す。

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比較例

不純物として Fe を含む溶融 Al-IC、Fe の 1.0 ~ 6.0 倍量に相当する Mn を含む Al-Mn 合金を 850°C で添加して溶融し、冷却して 665°C で 80 分間保持した後、晶出した金属間化合物を沪過分離した。ここで晶析前及び晶析分離後ににおける溶融 Al 中の Fe 及び Mn の含有率を測定し、第 1 表の結果を得た。尚この結果は第 2 図の ○ → ● に相当する。

第 1 表

| 晶析前    |        | 晶析分離法  |        | 保持温度 |
|--------|--------|--------|--------|------|
| Fe (%) | Mn (%) | Fe (%) | Mn (%) | (°C) |
| 1.9    | 1.9    | 1.1    | 1.1    | 665  |
| 1.6    | 3.1    | 0.7    | 1.4    | "    |
| 0.5    | 3.0    | 0.8    | 1.7    | "    |

この結果からも明らかな如く、Fe の含有率を相当低下させることはできるが、その効果は尚不

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第 2 表

| 晶析前 (%) |      |      | 晶析分離後 (%) |      |      | 保持温度 |
|---------|------|------|-----------|------|------|------|
| Fe      | Mn   | Mg   | Fe        | Mn   | Mg   | (°C) |
| 0.75    | 2.50 | 3.80 | 0.85      | 1.20 | 3.4  | 645  |
| 1.10    | 1.80 | 3.1  | 0.70      | 0.90 | 3.5  | "    |
| 0.50    | 2.50 | 6.1  | 0.18      | 0.90 | 6.7  | 680  |
| 0.80    | 2.10 | 6.2  | 0.85      | 0.75 | 6.8  | "    |
| 0.85    | 1.15 | 6.0  | 0.55      | 0.75 | 6.7  | "    |
| 0.71    | 2.26 | 9.8  | 0.29      | 0.71 | 10.8 | 590  |
| 2.08    | 1.01 | 10.1 | 0.96      | 0.45 | 12.0 | "    |
| 0.94    | 1.71 | 20.5 | 0.86      | 0.49 | 26.1 | 520  |
| 1.60    | 1.12 | 18.9 | 0.68      | 0.88 | 25.7 | "    |

この結果からも明らかな様に、Mg を添加することにより、Mg 無添加の場合（比較例）に比べて Fe 及び Mn の含有率を相当低下させ得ることが確認できる。

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満足であり、しかも相当多量の Mn が混入している。

実施例 1

不純物として Fe を含む溶融 Al-IC、Fe の 1.5 ~ 5.0 倍量に相当する Mn を含む Al-10% Mn 合金を 850°C で添加して溶融し、更に 750°C に降温して Mg 濃度が 8%、6%、10% 及び 20% となる様に Al-80% Mg 合金を添加し、夫々を 645°C 又は 680°C、590°C、520°C で 80 分間保持した後、晶出した金属間化合物を沪過分離した。ここで晶析前及び晶析分離後ににおける溶融 Al 中の Fe、Mn 及び Mg の含有率を測定し、第 2 表の結果を得た。尚この結果は第 2 図の △ → ▲、◇ → ◆、☆ → ★ 及び ○ → ● に相当する。

実施例 2

不純物として Fe 及び Cr を含む溶融 Al-IC に対し、これら不純物の 1.15 ~ 1.22 倍量に相当する Al-10% Mn を 850°C で添加して溶融し、更に 750°C に降温した後 Mg 濃度が 6% となるよう金属 Mg を加えて溶融し、680°C で 80 分間保持する。その後晶出した金属間化合物を沪過分離し、晶析分離の前・後ににおける溶融 Al 中の各金属不純物の含有率を比較した。結果を第 3 表に示す。

第 3 表

|    | 晶析前            | 晶析分離後          |
|----|----------------|----------------|
| Fe | 1.9 1<br>1.8 9 | 1.1 0<br>1.0 8 |
| Mn | 1.8 9          | 1.0 9<br>1.0 7 |
| Mg | 6.1 0<br>6.0 0 | 7.2 0<br>7.0 0 |
| Cr | 0.2 9<br>0.4 1 | 0.2 0<br>0.1 9 |

（単位：重量%・以下同様）

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## 実施例 3

不純物として Fe 及び Mn を含む溶融 Al IC に対し、これら不純物の 1.08、1.12 倍量に相当する Al - 10% Mn を 850°C で添加して溶融し、更に 750°C で降温した後 Mg 濃度が 6% となるように金属 Mg を加えて溶融し、以下実施例 2 と同様にして晶出した金属間化合物を汎過分離し、晶析分離前・後ににおける溶融 Al 中の金属不純物の含有率を比較した。結果を第 4 表に示す。

第 4 表

|    | 晶析前  | 晶析分離後 |
|----|------|-------|
| Fe | 1.90 | 1.21  |
|    | 1.90 | 1.12  |
| Mn | 1.92 | 1.18  |
|    | 1.90 | 1.12  |
| Mg | 6.00 | 6.80  |
|    | 5.80 | 6.90  |
| Mo | 0.05 | 0.08  |
|    | 0.28 | 0.08  |

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## 実施例 5

不純物として Fe 及び As を含む溶融 Al に対し、これら不純物の 1.01 倍量に相当する Al - 10% Mn を 850°C で添加し、以下実施例 8 と同様に 6% 相当の金属 Mg を液加・溶融して晶出した金属間化合物を汎過分離し、晶析分離前・後ににおける溶融 Al 中の金属不純物の含有率を比較した。結果を第 6 表に示す。

第 6 表

|    | 晶析前  | 晶析分離後 |
|----|------|-------|
| Fe | 1.89 | 1.10  |
|    | 1.90 | 1.12  |
| Mn | 1.87 | 1.15  |
|    | 1.92 | 1.12  |
| Mg | 6.10 | 6.80  |
|    | 5.80 | 6.90  |
| As | 0.02 | 0.01  |
|    | 0.04 | 0.01  |

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## 実施例 4

不純物として Fe 及び Pb を含む溶融 Al IC に対し、これら不純物の 1.12、1.34 倍量に相当する Al - 10% Mn を 850°C で添加し、以下実施例 8 と同様に 6% 相当の金属 Mg を添加・溶融して晶出した金属間化合物を汎過分離し、晶析分離前・後ににおける溶融 Al 中の金属不純物の含有率を比較した。結果を第 5 表に示す。

第 5 表

|    | 晶析前  | 晶析分離後 |
|----|------|-------|
| Fe | 1.92 | 1.15  |
|    | 1.98 | 1.20  |
| Mn | 1.89 | 1.08  |
|    | 1.90 | 1.10  |
| Mg | 6.00 | 7.20  |
|    | 5.70 | 7.10  |
| Pb | 0.28 | 0.18  |
|    | 0.65 | 0.51  |

第 8 ~ 6 表の結果からも明らかに如く、本発明は Fe のみならず Cr、Mo、Pb、As 等の不純物についても相当の除去効果を發揮することが理解される。

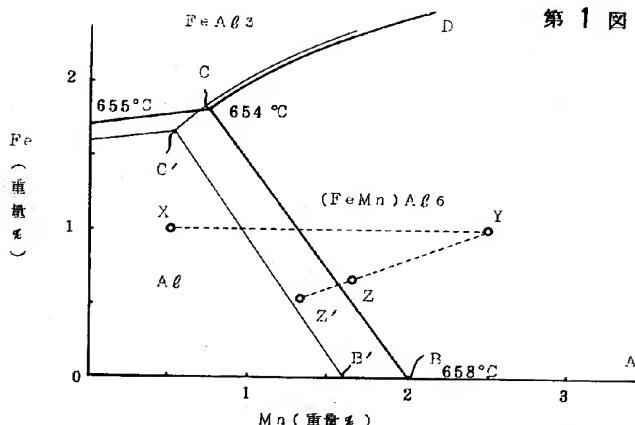
## 4. 図面の簡単な説明

第 1 図は Al - Mn - Fe 系及びこれに Mg が介在したときの 3 元状態図、第 2 図は Mg の添加効果を例示するグラフ、第 3 図は Mg が Al - Mn - Fe 系の共晶温度（融点）に及ぼす影響を示すグラフ、第 4 図は Mn / Fe 除去率の関係を示すグラフ、第 5 図は Mg の添加率と Fe 除去率の関係を示すグラフである。

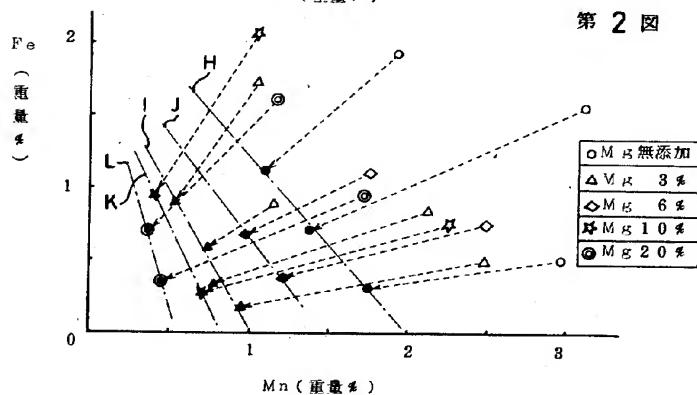
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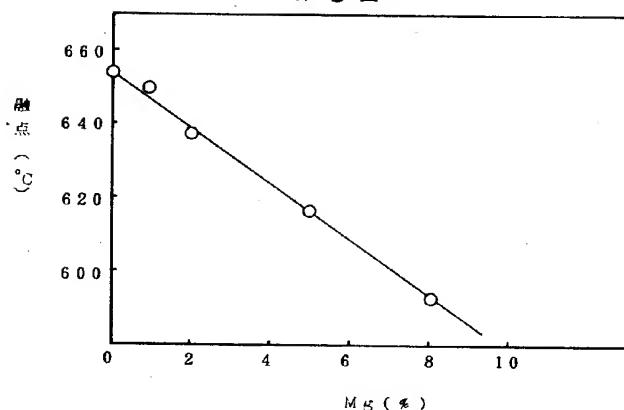
第1図



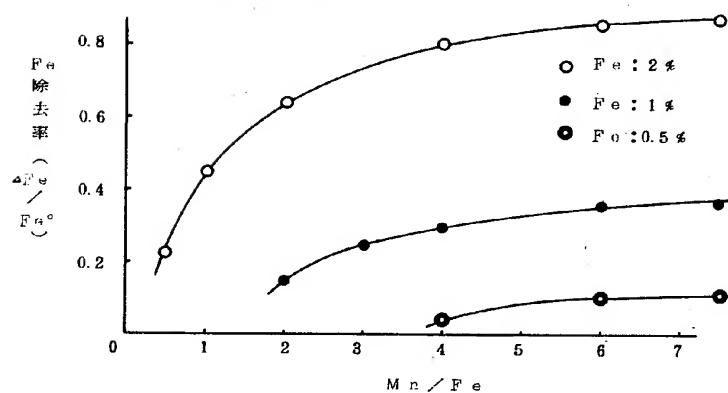
第2図



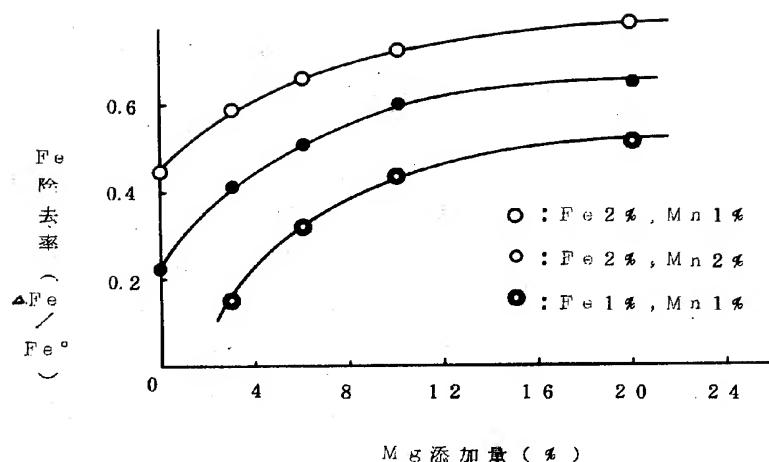
第3図



第4図



第5図



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REFINEMENT METHOD OF ALUMINUM OR ALUMINUM ALLOY  
[Aruminyumu matawa aruminyumu gokin no seizo hoho]

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Claim

A refinement method of aluminum or aluminum alloy characterized by the following facts: in a melt of aluminum or aluminum alloy containing metal impurities, metal manganese and/or aluminum-manganese alloy in quantity 0.5-6 times that of the metal impurities contained in the melt and metal magnesium and/or aluminum-magnesium alloy in quantity 0.002-0.1 times that of the melt are added, melted and mixed; the melt mixture is cooled to a temperature ranging from the eutectic

temperature to the eutectic temperature plus 50°C and is kept at said temperature, so that the metal impurities are crystallized and separated as intermetallic compounds.

Detailed explanation of the invention

The present invention pertains to a refinement method of aluminum or aluminum alloy (hereinafter to be referred to as Al). More specifically, the present invention pertains to a refinement method of Al characterized by the fact that it can efficiently remove the metal impurities, such as iron as well as chromium, molybdenum, lead, arsenic, etc., contained in Al.

In the recent years, in the aluminum industry, people have been engaged in development of schemes for effective use of Al scraps so as to conserve resources and to stabilize the metal price. However, the Al scraps contain iron and a lot of other types of metal impurities. As there is yet no refinement technology, while some can be used as the feed material for casting alloy, most of them cannot be used as feed material for rolling alloy that has a strict requirement on components.

People have performed extensive studies on the refinement method of Al, and have proposed the following schemes: {a} 3-layer type electrolysis refinement method in which the metal impurities in Al are separated by means of electrolysis; {b} method in which the metal impurities, especially iron, are separated as Al-silicon-iron intermetallic compounds; {c} method in which Al-manganese alloy with a higher melting point is added in the Al melt so that the metal impurities, especially iron, are adsorbed on the solid phase alloy, and can be removed as intermetallic compounds with Al-manganese alloy, etc. However, the aforementioned prior art has some problems. For said method {a}, while high purity Al can be obtained, the refinement operation is nevertheless complicated and the refinement fee is extremely high. Consequently, it is inappropriate as the refinement processing method of the scraps. For method {b}, a large quantity of silicon is mixed in Al during the refinement operation, so that its

application is limited to extremely special type of Al-silicon alloys. On the other hand, for said method {c}, the refinement operation is simple and iron can be reduced significantly, and, even when a small quantity of manganese is mixed in Al, the impurities effect is little displayed, so that it is the best method from the industrial viewpoint. However, in this method, as iron is adsorbed and removed due to the solid-liquid reaction, the frequency of contact between iron and Al-manganese alloy becomes lower, and the reaction rate becomes very low. Consequently, the removal rate of iron is significantly lower than the other methods, and it has little value of application as the refinement method of Al with a strict requirement on its composition.

In consideration of the aforementioned problems, the present inventors have performed research on the scheme for increasing the impurities removal efficiency by increasing the contact frequency with the metal impurities and the reaction rate as contact is made between Al-manganese alloy and Al in melt state. As a result of the research performed on solving the problem of said method {c} that although the removal efficiency is improved significantly, it is still not satisfactory, it was found that if an appropriate quantity of magnesium is added together with manganese in the Al for refinement, and the obtained melt mixture is cooled to a temperature a little higher than the eutectic temperature, almost all of the metal impurities can be crystallized and separated as intermetallic compounds with Al-manganese alloy. Also, in this process, in addition to iron, other impurities, such as chromium, molybdenum, lead, arsenic, etc., also can be removed efficiently. As a result, the present invention was reached.

That is, the present invention provides a refinement method of aluminum or aluminum alloy characterized by the following facts: in a melt of aluminum or aluminum alloy containing metal impurities, metal manganese and/or aluminum-manganese alloy in quantity 0.5-6 times that of the metal impurities contained in the melt and metal magnesium and/or aluminum-magnesium alloy in quantity 0.002-0.1 times that of the melt are added, melted and mixed; the melt mixture is cooled to a

temperature ranging from the eutectic temperature to the eutectic temperature plus 50°C and is kept at said temperature, so that the metal impurities are crystallized and separated as intermetallic compounds.

In the following, an explanation will be given in more detail regarding the constitution and operation of the present invention with reference to figures. However, the present invention is not limited to the following typical examples, and appropriate changes are allowed as long as the gist of the present invention is observed. They are also included in the technical range of the present invention.

Figure 1 is a three-dimensional state diagram illustrating the Al-manganese (Mn)-iron (Fe) system. In region ABCD, intermetallic compound  $(\text{FeMn})\text{Al}_6$  is formed as the initial crystal. Here, for the melt mixture with the composition shown at point X, after Al-Mn alloy with composition shown at point Y is added and fully melted, the mixture is cooled to a temperature in the range lower than the liquid phase temperature and higher than the eutectic temperature, and it is then kept at this temperature, a intermetallic compound  $(\text{FeMn})\text{Al}_6$  is crystallized, and the melt is changed to the composition indicated by point Z. Consequently, by separating the crystallized substance, it is possible to obtain a melt Al alloy with a low Fe concentration.

However, in this method, removal of Fe is still insufficient, and the quantity of Mn melted and dissolved in Al alloy is significant. However, if Al-Mn alloy is added together with addition of the Al-Mn alloy or during the cooling process of the melt after Al-Mn alloy is completely melted, the three-dimensional state diagram changes to the state indicated by the broken line in Figure 1, and the crystallization region of  $(\text{Fe, Mn})\text{Al}_6$  spreads to region AB'C'D' on the lower temperature side. That is, by having Al-Mg alloy coexisting in the melt mixture, the eutectic temperature falls, and  $(\text{Fe, Mn})\text{Al}_6$  can be crystallized at an even lower temperature. Consequently, point Z may move to point Z', and, by separating and removing the crystallized substance in the same way, it is possible to significantly increase the removal rate for Fe and Mn in Al.

Figure 2 is a diagram illustrating another example of the effect for adding Mg. In this graph, the contents of Fe and Mn in the Al alloy before and after separation and removal of the crystallized substance are shown. As can be seen from such results, when Mg is not added, reduction in the contents of Fe and Mn is limited to the quantity corresponding to H-line (corresponding to B-C line in Figure 1). On the other hand, when 3% of Mg is added, it is possible to reduce the contents of Fe and Mn to the quantity corresponding to I-line; when 6% of Mg is added, it is possible to reduce the contents of Fe and Mn to the quantity corresponding to J-line; when 10% of Mg is added, it is possible to reduce the contents of Fe and Mn to the quantity corresponding to K-line; and when 20% of Mg is added, it is possible to reduce the contents of Fe and Mn to the quantity corresponding to L-line. In this way, according to the present invention, by adding Mg together with Mn in the Al melt containing iron as the impurity, and by increasing the quantity of Mg added, it is possible to significantly reduce the contents of Fe and Mn. As to be shown below in the application examples, according to the present invention, in addition to Fe, it is also possible significantly reduce the other metal impurities, such as chromium, molybdenum, lead, arsenic, etc.

In addition, Figure 3 is a diagram illustrating the results of the measurement of the relationship between the Mg quantity added and the eutectic temperature (melting point) for 95% Al - 2% Mn - 2% Fe. The excellent effect of Mg in lowering the melting point of Al under treatment can be clearly seen.

As far as Mn used in the present invention is concerned, either metal Mn or Al-Mn alloy may be used. In order to ensure that it can be melted quickly after it is added in Al, it is preferred that Al-Mn alloy containing about 5-30% of Mn be added. Also, as far as the temperature of the Al melt when Mn is added is concerned, in consideration of the melting rate, a temperature range of 720°C - 850°C is most preferred. In this case, the quantity of Mn added as pure Mn content should be selected in the range of 0.5-6.0 times the quantity of Fe contained in the Al melt under treatment. If the Mn quantity is less than

the aforementioned range, the quantity of the intermetallic compound becomes insufficient, and Fe and other metal impurities cannot be well removed. On the other hand, if the Mn quantity is more than the aforementioned range, as can be seen from Figure 4, there is little increase in the impurities removal rate. Also, the quantity of the crystallized  $(\text{Fe}, \text{Mn})\text{Al}_6$  is increased, so that the quantity of the Al refined product is reduced, and the yield falls. Consequently, this is undesired for practical application.

Also, as far as Mg is concerned, either metal Mg or Al-Mg alloy may be used. The temperature for adding it is a little lower than the temperature for adding Al-Mn alloy (the temperature for facilitating melting of Al-Mg alloy in Al melt), that is, in the range of 700-800°C. Here, the quantity of Mg added should be selected in the range of 0.002-0.1 times the total quantity of the Al melt to be treated. If the quantity is smaller than the aforementioned range, the effect in lowering the eutectic temperature is little displayed, and Fe and other metal impurities and the content of Mn cannot be sufficiently reduced. On the other hand, if the quantity is over the aforementioned range, as shown in Figure 5, the removal rate for the metal impurities, such as Fe, Mn, etc., becomes not so significant, and the purpose of the present invention cannot be realized.

In this way, by adding an appropriate quantity of Mn (or Al-Mn alloy) and Mg (or Al-Mg alloy) in the Al melt and having them melted and blended and then cooling the melt mixture slowly,  $(\text{Fe}, \text{Mn})\text{Al}_6$  can be crystallized. Here, if the temperature of the melt is higher than the liquid phase line temperature, said  $(\text{Fe}, \text{Mn})\text{Al}_6$  is not crystallized. On the other hand, if the temperature is lower than the eutectic temperature, together with  $(\text{Fe}, \text{Mn})\text{Al}_6$ , pure Al is also crystallized, so that it is impossible to have  $(\text{Fe}, \text{Mn})\text{Al}_6$  alone separated. Consequently, although the crystallization temperature should be higher than the eutectic temperature and lower than the liquid phase line temperature, the present inventors have performed extensive studies on the crystallization temperature for increasing the removal rate of the metal impurities to the maximum limit. As a result, it has been found that the most appropriate

temperature range is up to 50°C higher than the eutectic temperature. By keeping within this temperature range, crystallization of (Fe, Mn)Al<sub>6</sub> can be performed at the highest efficiency.

There is no specific restriction on the means for separating the crystallized intermetallic compound from the melt Al. One may adopt the conventional methods or the solid-liquid separating methods to be developed in the future. Examples of the specific methods that may be adopted include the precipitation separation method that makes use of the fact that the specific gravity of the intermetallic compound is higher than the specific gravity of the melt Al, filtering separating method by means of a porous material, centrifugal separating method, etc.

According to the present invention with the constitution described schematically above, appropriate quantities of Mn and/or Al-Mn and Mg and/or Al-Mg are added in the Al melt containing the metal impurities, and the intermetallic compound is crystallized at a prescribed temperature. As a result, it is possible to make efficient removal of Fe and other impurities, such as chromium, molybdenum, lead, arsenic, etc., in the Al melt, so that high purity Al or Al alloy can be recovered. Consequently, the present invention has a very high practical application value as a refinement method for various types of impure Al and Al alloys, such as Al scraps.

In the following, the present invention will be explained in more detail with reference to application examples.

#### Comparative example

In melt Al containing Fe as the impurity, Al-Mn alloy containing Mn in quantity corresponding to 1.0-6.0 times the quantity of Fe was added and melted, followed by cooling to 665°C and then kept at this temperature for 30 min. Then, the crystallized intermetallic compound was filtered and separated.

Here, the contents of Fe and Mn in the melt Al were measured before and after the crystallization and separation, with results listed in Table 1. The results correspond to O → • in Figure 2.

Table 1

| 分析前    | 分析後    | 保持温度  |        |      |
|--------|--------|-------|--------|------|
| Fe (%) | Mn (%) | P (%) | Mn (%) | (°C) |
| 1.9    | 1.9    | 1.1   | 1.1    | 855  |
| 1.8    | 3.1    | 0.7   | 2.4    | —    |
| 0.5    | 3.0    | 0.8   | 2.7    | —    |

Key: 1 Before crystallization

After crystallization and separation

Holding temperature

As can be seen from these results, although the content of Fe is significantly reduced, the effect is still insufficient, and a significant quantity of Mn is mixed in.

#### Application Example 1

At 850°C, Al-10% Mn alloy containing Mn in a quantity corresponding to 1.35-5.0 times the quantity of Fe was added in melt Al containing Fe as impurity, and the melt mixture was cooled to 750°C. Then, Al-80% Mg alloy was added to have Mg concentration of 3%, 6%, 10% and 20%, respectively. After being kept at 645°C, 630°C, 590°C, and 520°C, respectively, for 30 min, the crystallized intermetallic compound was filtered and separated. Here, before crystallization and after crystallization and separation, the contents of Fe, Mn and Mg in the melt Al were measured, with the results listed in Table 2. Here, the results correspond to △→▲, ◇→◆, ★→★ and ○→●, respectively.

Table 2

| Al-Mn-Mg |      |      | 分析分離後 |      |      | 保持温度<br>(°C) |
|----------|------|------|-------|------|------|--------------|
| Fe       | Mn   | Mg   | Fe    | Mn   | Mg   |              |
| 0.75     | 2.54 | 3.60 | 0.84  | 1.80 | 3.4  | 643          |
| 1.10     | 1.59 | 2.1  | 0.76  | 0.90 | 1.8  | —            |
| 0.50     | 2.50 | 0.1  | 0.52  | 0.90 | 6.7  | 680          |
| 0.80     | 2.10 | 0.22 | 0.86  | 0.76 | 6.8  | —            |
| 0.85     | 1.15 | 0.0  | 0.55  | 0.75 | 6.7  | —            |
| 0.71     | 2.26 | 0.8  | 0.29  | 0.71 | 1.08 | 690          |
| 2.02     | 1.01 | 1.01 | 0.94  | 0.45 | 1.25 | —            |
| 0.94     | 1.71 | 2.05 | 0.85  | 0.49 | 2.41 | 620          |
| 1.68     | 1.12 | 1.89 | 0.59  | 0.58 | 2.57 | —            |

Key: 1 Before crystallization

After crystallization and separation

Holding temperature

As can be seen from these results, by adding Mg, the contents of Fe and Mn can be reduced to a level much lower than the case when Mg is not added (comparative example).

### Application Example 2

At 850°C, Al-10% Mn alloy containing Mn in a quantity corresponding to 1.15-1.22 times the quantity of Fe and Cr as impurities was added in melt Al, and the melt mixture was cooled to 750°C. Then, Mg metal was added to have Mg concentration of 6%. After being kept at 680°C for 30 min, the crystallized intermetallic compound was filtered and separated. Here, before crystallization and after crystallization and separation, the contents of the metal impurities in melt Al were measured, with the results listed in Table 3.

Table 3

|    | 晶界部  | 析出物  |
|----|------|------|
| Fe | 3.8% | 1.1% |
| Mn | 1.1% | 1.0% |
| Mo | 8.1% | 3.8% |
| C  | 0.2% | 0.3% |

(单位:质量% - GOFUN)

Key: 1 Before crystallization

After crystallization and separation

2 (Units: wt%, same in the following)

Application Example 3

At 850°C, Al-10% Mn alloy containing Mn in a quantity corresponding to 1.08 and 1.12 times the quantity of Fe and Mo as impurities was added in melt Al, and the melt mixture was cooled to 750°C. Then, metal Mg was added to have a Mg concentration of 6%. Then, in the same way as in Application Example 2, the crystallized intermetallic compound was filtered and separated. Here, before crystallization and after crystallization and separation, the contents of the metal impurities in melt Al were measured, with the results listed in Table 4.

Table 4

(1)

|     | 前 置 后          | 后置分离法          |
|-----|----------------|----------------|
| P * | 1.9 0<br>1.9 0 | 1.2 1<br>1.1 2 |
| M n | 1.9 2<br>1.9 0 | 1.1 3<br>1.1 2 |
| M g | 6.0 0<br>5.8 0 | 6.8 0<br>6.9 0 |
| M o | 0.0 2<br>0.3 5 | 0.0 8<br>0.0 8 |

Key: 1 Before crystallization

After crystallization and separation

#### Application Example 4

At 850°C, Al-10% Mn alloy containing Mn in a quantity corresponding to 1.12 and 1.34 times the quantity of Fe and Pb as impurities was added in melt Al. Then, in the same way as in Application Example 3, metal Mg was added to have a Mg concentration of 6%, and the crystallized intermetallic compound was filtered and separated. Here, before crystallization and after crystallization and separation, the contents of the metal impurities in melt Al were measured, with the results listed in Table 5.

Table 5

|     | 前 置 后          | 后置分离法          |
|-----|----------------|----------------|
| P * | 1.9 2<br>1.9 0 | 1.1 3<br>1.2 0 |
| M n | 1.5 2<br>1.9 0 | 1.0 3<br>1.1 0 |
| M g | 6.0 0<br>5.7 0 | 7.2 0<br>7.1 0 |
| P b | 0.2 2<br>0.3 5 | 0.1 8<br>0.6 1 |

Key: 1 Before crystallization

After crystallization and separation

Application Example 5

At 850°C, Al-10% Mn alloy containing Mn in a quantity corresponding to 1.01 times the quantity of Fe and As as impurities was added in melt Al. Then, in the same way as in Application Example 3, metal Mg was added to have a Mg concentration of 6%, and the crystallized intermetallic compound was filtered and separated. Here, before crystallization and after crystallization and separation, the contents of the metal impurities in melt Al were measured, with the results listed in Table 6.

Table 6

|    | 前<br>行<br>能  | 后<br>行<br>能  |
|----|--------------|--------------|
| Fe | 1.89<br>1.90 | 1.10<br>1.12 |
| Mn | 2.87<br>2.92 | 1.16<br>1.12 |
| As | 0.10<br>0.09 | 6.89<br>6.94 |
| Al | 0.02<br>0.04 | 0.01<br>0.02 |

Key: 1 Before crystallization

After crystallization and separation

As can be seen from the results listed in Tables 3-6, according to the present invention, not only Fe, but also Cr, Mo, Pb, As, and other impurities can be significantly removed.

Brief explanation of figures

Figure 1 is a three-dimensional state diagram illustrating the state of the Al-Mn-Fe system and when Mg is included in it. Figure 2 is a graph illustrating the effect of addition of Mg. Figure 3 is a graph

illustrating the influence of Mg on the eutectic temperature (melting point) of the Al-Mn-Fe system.

Figure 4 is a graph illustrating the relationship of the Mn/Fe removal rate. Figure 5 is a graph illustrating the relationship between the quantity of Mg added and the Fe removal rate.

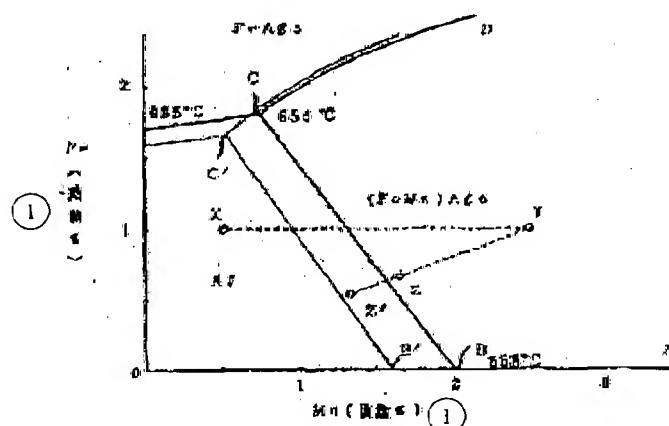


Figure 1

Key: 1 (wt%)

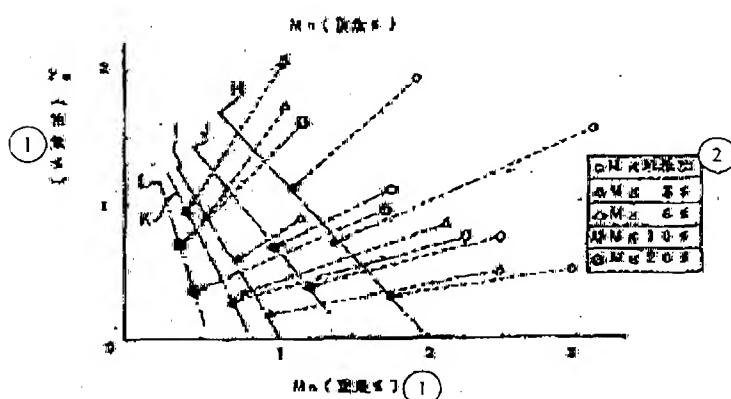


Figure 2

Key: 1 (wt%)

2 Mg not added

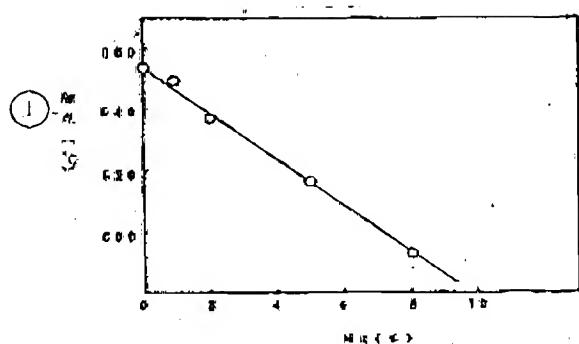


Figure 3

Key: 1 Melting point

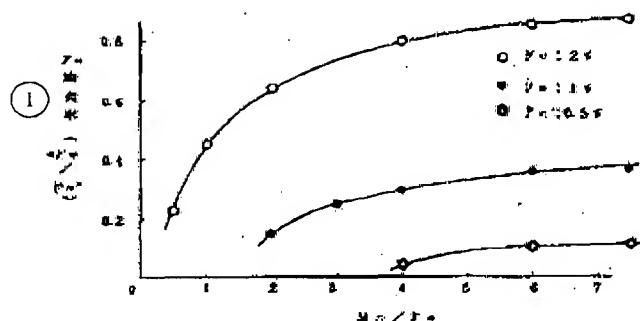


Figure 4

Key: 1 Fe removal rate

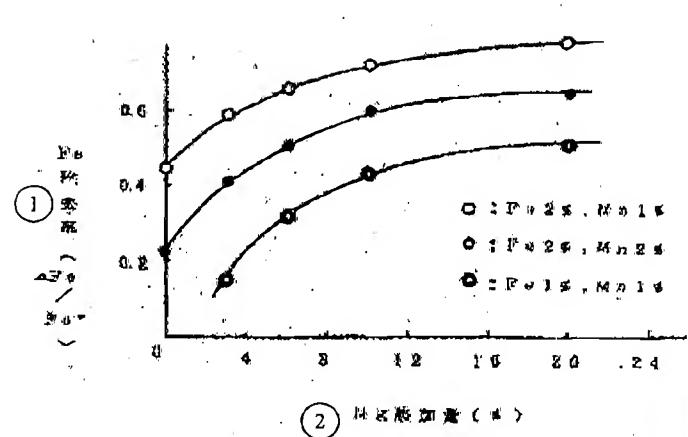


Figure 5

Key: 1 Fe removal rate

2 Quantity of Mg added